Concentration of Charge Carriers and Anomalous Gap Parameter in the Normal State of High-T_c Superconductors

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Fermi-Dirac statistics has been utilized by introducing the average ionization energy (E_I) as an additional anomalous energy gap in order to derive the two-dimensional concentration of charge carriers and the phenomenological resistivity model for the superconducting polycrystalline materials. The best fitted values of E_I and the charge carriers' concentration ranges in the vicinity of 4 to 9 meV and 10^{16} m^{-2} respectively for the superconducting single crystal samples and polycrystalline compounds synthesized with various compositions via solid-state reactions. The phenomenological resistivity model is further redefined here based on the gapless nature of charge-carriers' dynamics within the Cu-O$_2$ planes that corresponds to anomalous Fermi liquid behavior, which is in accordance with the nested Fermi liquid theory.

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I. INTRODUCTION

Since the reported discovery of high-T_c superconductors (HTSC) based on the copper oxide materials, the scientific communities have been swept with revolutionary excitement. This leads to intensified focus on this particular material both in term of theoretical and experimental research throughout the world. The discovered Ba-La-Cu-O system by Bednorz and Muller [1] with a T_c of 33 K have had led to subsequent findings of superconductivity in La-Sr-Cu-O at 40 K [2,3] and later in Y-Ba-Cu-O system at 90 K [4]. Actually the La-Ca,Sr,Ba-Cu system were first synthesized and studied by Raveau et al. [5]. The conduction mechanisms in the normal state of HTSC such as the out-of-plane and in-plane resistivities are intriguing due to the highly anisotropic behavior. Anisotropic normal state electrical properties such as metallic-like conduction in the Cu-O$_2$ planes and semiconducting-like conduction between those planes [6] are believed to be one of the central concerns for applications [7]. This in turn may lead one to extract some clues to understand the mechanism of high-T_c superconductivity [7,8]. Interestingly, Levin et al. [9] using the T-dependence on anisotropy ρ_c/ρ_{ab} showed the conductivity relationship between σ_c and σ_{ab} as: $\sigma_{ab} = (a + bT^{-2/3})\sigma_c$. This $T^{-2/3}$ dependence point towards a 2-dimensional (2D) localization and hopping. Quite recently, Basov et al. [10] have analyzed the interlayer infrared conductivity of HTSC. They found that discrepancy occurred between superfluid density, ρ_s and difference between normal state (N_n) and superconducting state (N_s) spectral weights i.e. $\rho_s > N_n - N_s$. This discrepancy was only revealed for c-axis electrodynamics but could not find a similar conflict in ab-plane ($\rho_s = N_n - N_s$), which can be interpreted as a gapless behavior of charge carriers in ab-planes. PrBa$_2$Cu$_3$O$_7$ compound gives a unique non-superconducting, semiconducting-like normal state electrical property and the nature of this unique property was discussed intensively in terms of hole doping in Cu-O$_2$ planes [11–13]. As a matter of fact, semiconducting-like behavior of $\rho(T)$ corresponds to localization of charge carriers connected with oxygen disorder in Cu-O$_x$ chains and the disorder in Pr^{3+}, Pr^{4+} distribution [14]. Furthermore, positron annihilation studies [15,16] showed that PrBa$_2$Cu$_3$O$_7$ compound has the Fermi surface due to Cu-O$_x$ chains' state. The concept of charge concentration in high-T_c cuprates is very ambiguous [17]. However, it is well established that T_c of cuprates depends on holes' concentration within Cu-O$_2$ planes where carrier concentration is determined by counting the valence number [17,18]. Very recently, concentration of interlayer electrons and its resistivity model for HTSC was derived via Fermi-Dirac statistics with an additional gap constraint, which is the average ionization energy (E_I) [19]. In this letter however, a phenomenological polycrystalline resistivity model is derived based on E_I as well and is redefined here to obtain an ab-plane resistivity model by incorporating gapless nature of charge carriers in Cu-O$_2$ planes. Besides that, estimation for concentration of charge carriers and E_I via best fitting in the normal state of HTSC are also illustrated in this work. Apart from that, estimation of resistivity transition such as from metallic to semiconductor or vice versa upon substitution in crystalline 123 phase Pr$_x$Y$_{1-x}$Ba$_2$Cu$_3$O$_7$ system is also highlighted. Ions' average ionization energies were calculated from ref. [20].

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II. CHARGE CARRIERS' CONCENTRATION AND RESISTIVITY MODELS

Tunneling electron's distribution has been derived via Fermi-Dirac (FD) statistics using average ionization energy (E_I) as an anomalous constraint, and is given below [19]

$$f_e(E) = exp[-\mu - \lambda(E + E_I)]. \tag{1}$$

Similarly, probability function for holes can be derived and is given in Eq. 2 below, where it is obtained by inserting the restrictive condition $E_{hole} = E_{initialstate} - E_I$. $E_{initialstate}$ is the energy of a particle in a given system at a certain initial state and ranges from 0 to $-\infty$. This can be justified as follows; for a hole to occupy a lower state M from initial state N is more probable than to occupy state L if the condition $E_I(M) < E_I(L)$ at certain temperature, T is satisfied.

$$f_h(E) = exp[\mu + \lambda(E - E_I)]. \tag{2}$$

Note that Eq. 1 and Eq. 2 can be reduced to the standard Fermi-Dirac distributions if E_I approaches 0. The importance of average E_I's inclusion is that, it can be interpreted as a c-axis gap, which represents the gap that tied to a neutral atom and could be varied with ionic substitutions. The respective normal state concentration of electrons [19] and holes are given by

$$n = \int_0^\infty f_e(E) N_e(E) dE, \tag{3}$$

$$p = \int_{-\infty}^0 f_h(E) N_h(E) dE. \tag{4}$$

$\mu = -E_F/k_B T$, $\lambda = 1/k_B T$ are carefully chosen to confine the total number of particles in a given system equals to n. $N_e(E)$ and $N_h(E)$ are 2D density of states (DOS) that can be derived from the Schrodinger equation. $N_e(E)$ and $N_h(E)$ are derived as $m_e^*/\pi\hbar^2$ and $m_h^*/\pi\hbar^2$ respectively and note that both 2D DOS are independent of E. Solutions of integral Eq. 3 and Eq. 4 give the concentration of tunneling electrons between Cu-O_2 layers, n and concentration of holes, p in the normal state of HTSC as

$$n = \frac{m_e^* k_B T}{\pi\hbar^2} exp\left[\frac{E_F - E_I}{k_B T}\right]. \tag{5}$$

$$p = \frac{m_h^* k_B T}{\pi\hbar^2} exp\left[\frac{-E_F - E_I}{k_B T}\right]. \tag{6}$$

Eq. 5 and Eq. 6 are expressed in term of ionization energy, where k_B is Boltzmann constant, m_e^* and m_h^* are electrons' and holes' effective masses respectively, $\hbar =$ $h/2\pi$, h is Plank constant and E_F is Fermi level. Moreover, from Eq. 5 and Eq. 6, concentration of charge carriers in the normal state of polycrystalline HTSC can be written in the form of geometric mean which is given by

$$\sqrt{np} = \frac{k_B T}{\pi\hbar^2} \sqrt{m_e^* m_h^*} exp\left[\frac{-E_I}{k_B T}\right]. \tag{7}$$

This equation is free from Fermi level, hence concentration of charge carriers in superconducting polycrystals can be calculated by first estimating E_I, which is described in the following paragraph. Scattering among fermions leads to the conclusion of $1/\tau \propto T^2$ [6] and has been confirmed empirically that $\rho \propto T^2$ as well [21–23]. Furthermore, by utilizing metallic resistivity equation of $\rho = m/ne^2\tau$ where e and τ are electron's charge and mean scattering free time respectively [24] then one can derive the normal state phenomenological model for HTSC as given in Eq. 8 below. This were carried out by substituting $m = m_e^* = m_h^*$, $n = \sqrt{np}$, and $1/\tau = AT^2$ into metallic resistivity equation.

$$\rho_{poly}(T, E_I) = A\frac{\pi\hbar^2}{e^2 k_B} T exp\left[\frac{E_I}{k_B T}\right]. \tag{8}$$

A is independent of temperature. Eq. 8 above can be used in the normal state resistivity of polycrystalline superconducting samples to evaluate the variations in E_I. Substituting this E_I into Eq. 7 at certain temperature, T leads to the estimation of charge carriers' concentration. Subsequently, an ab-plane resistivity model can be derived quite naturally by taking anomalous gap = 0 due to gapless behavior of charge carriers in Cu-O_2 planes [10], thus Eq. 8 can be simply rewritten as

$$\rho_{ab}(T) = A\frac{\pi\hbar^2}{e^2 k_B} T. \tag{9}$$

This latter phenomenon could be due to change in the scattering rate that results from a different Fermi surface phase space argument in accordance with the nested Fermi liquid theory [25].

III. DISCUSSION

Polycrystalline resistivities, $\rho_{poly}(T)$ of crystalline YBa$_2$Cu$_3$O$_7$ [26] samples (B and C) were obtained using

$$\rho_{poly}(T) = \sqrt{\rho_c(T)\rho_{ab}(T)}. \tag{10}$$

It is assumed that Eq. 10 is valid to convert $\rho_c(T)$ and $\rho_{ab}(T)$ of single crystals to $\rho_{poly}(T)$ in order to analyze the effect or changes of A parameter between single crystals and polycrystals. Consequently, one can study the scattering effect solely on A parameter. Fitting of experimental $\rho_{poly}(T)$ with Eq. 8 gives respective values for

anomalous gap and concentration of charge carriers in the order of 4-9 meV and 10^{16} m^{-2} as listed in Table I. These fittings are depicted in Fig. 1 and Fig. 2 for samples B and C respectively. Furthermore, resistivity curves of polycrystalline Ga1212 [27], $Y_{1-x}Pr_xSrBaCu_3O_7$ [13], $YBa_2Cu_3O_{7-x}$ [28,29] and Tl1212 [30] samples are utilized by calculating the slope, $d\rho(T)/dT$ in the normal state of respective samples. This slope would enable one to determine E_I via Eq. 8 and followed by determination of concentration of charge carriers in the normal state at 300 K and 150 K, which are also listed in Table 1. The above concentration of charge carriers was estimated by substituting the best fitted E_I into Eq. 7. All samples have charge carriers' concentration in the order of 10^{16} m^{-2}, however, parameters A and E_I vary accordingly with samples. Note that A is equals to $A\pi\hbar^2/e^2k_B$ for simplicity's sake. It is quite interesting to note that all Y123 samples including the single crystals (sample A and B) have values for E_I between 7.69 and 8.25 meV. Contrary to that, A parameter is approximately 100 times lower for single crystals (10^{-8}) than polycrystals (10^{-6}) where this could be due to the nature of scattering in crystalline samples. Note also that the magnitude of $\rho_{poly}(T)$ estimated from Eq. 10 for single crystals are lower as mentioned previously. Similarly, A and E_I parameters for 1212 phase compounds, $Tl(Sr_{1.6}Sm_{0.4})CaCu_2O_{7-x}$, $Tl(Sr_{1.6}Dy_{0.4})CaCu_2O_{7-x}$ [30] and $GaSr_2(Y_{0.6}Ca_{0.4})Cu_2O_7$ [27] are in the order of 10^{-5} and 5-6 meV respectively. Substitution effect in $Y_{1-x}Pr_xSrBaCu_3O_7$ system is found to be not in favor of superconductivity where, Pr substitution increases anomalous gap, E_I and A parameter. Parallel to this, comparison between polycrystalline samples, $YBa_2Cu_3O_{7-x}$ (Sample A) [29] and $Y(Ba_{0.85}Pb_{0.15})_2Cu_3O_{7-x}$ (Sample B) [29] expose a reduction in E_I that suggests Pb substitution into Ba sites is somewhat favorable. Further comparisons between samples prepared by different listed researchers are not discussed here since the exact synthesis method varies from one researcher to another. In another context, $\rho_c(T)$ transition that occurs in superconducting $Y_{1-x}Pr_xBa_2Cu_3O_7$ system is in fact due to the differences in average E_I of $Pr^{3+,4+}$ and Y^{3+}. The valence number, $z+$ ($= 3+$) and $q+$ ($= 3+$) correspond to ions, Pr^{z+} and Y^{q+} respectively. Since E_I of Y^{3+} (1260 kJmol^{-1}) $> E_I$ of Pr^{3+} (1210 kJmol^{-1}) then substituting Y with Pr will reduce $E_I - E_F$ which is the c-axis gap parameter [19] for single crystals thus, reduces $\rho_c(T)$. Similar substitutions would also decrease the E_I gap parameter and $\rho_{poly}(T)$ which is for polycrystals that can be verified from Eq. 8. In contrast, the experimental data obtained by Jiang et al. (single crystals) [31] and Das et al. (polycrystals) [13] expose a contradicting relation where $\rho_c(T)$ and $\rho_{poly}(T)$ increases with Pr substitutions into Y sites. Thus valency of Pr is expected to be greater than 3+ due to Pr^{4+} ions' contribution [32]. Parallel to

this, Meng et al. [33] have employed PVL theory to determine valency of Pr in superconducting state. They have found that valency of Pr is vital in producing a superconducting $Y_{1-x}Pr_xBa_2Cu_3O_7$ compound. In their calculations, valency of Pr is < 3.15 to get a superconductor compound for all Pr doping but a valency of > 3.15 gives a superconducting state for only up to a limited Pr doping. Therefore, using the concept presented in section II, the minimum value of Pr's valence state, which is obviously above 3+ that contribute to non-metallic behavior, can be calculated from linear algebraic equation given below

$$\frac{\delta}{j}\sum_{i=z+1}^{z+j}E_{Ii} + \frac{1}{z}\sum_{i=1}^{z}E_{Ii} = \frac{1}{q}\sum_{i=1}^{q}E_{Ii} \qquad (11)$$

The first term, $\frac{\delta}{j}\sum_{i=z+1}^{z+j}E_{Ii}$ in Eq. 11 above has $i = z+1, z+2,..., z+j$ and $j = 1, 2, 3,....$. It is solely due to Pr^{4+} ions contribution or caused by reaction of the form $Pr^{3+} - electron \rightarrow Pr^{4+}$, hence j is equals to 1 in this case and δ represents the additional contribution from Pr^{4+}. The second ($i = 1, 2, 3, ..., z$) and last ($i = 1, 2, 3, ..., q$) terms respectively are due to reaction of the form $Pr - 3(electrons) \rightarrow Pr^{3+}$ and $Y - 3(electrons) \rightarrow Y^{3+}$. Recall that $q = z = 3+$ and $i = 1, 2, 3,...$ represent the first, second, third, ... ionization energies while $j = 1, 2, 3, ...$ represent the fourth, fifth, sixth, ... ionization energies. Therefore, $z + \delta$ gives the minimum valence number for Pr which is calculated to be 3.0133 from Eq. 11. This critical value corresponds specifically for $\rho(T)$ variations in the normal state behavior where $\rho(T)$ decreases with doping if and only if $z + \delta < 3.0133$ and vice versa if $z + \delta > 3.0133$. Those changes in $\rho(T)$ will eventually affect $d\rho(T)/dT$ slope transition in term of metallic-like or semiconducting-like normal state properties as mentioned for pure 1212 phase polycrystals [19]. In order to justify this critical value, $\rho_c(T)$ measurements of $Y_{1-x}Pr_xBa_2Cu_3O_7$ single crystals [31] that indicate a transition of $d\rho_c(T)/dT$ slope from positive to negative with x doping were used. Thus, from the above-presented approximation, $z + \delta$ or valence state of Pr for $x = 0.13$ to 0.42 samples is > 3.0133 since there is an increment of $\rho_c(T)$ with Pr doping. In contrast, $z + \delta$ value should be above 3.15 as mentioned previously in accordance with Meng et al. [33]. Although 3.15 is larger than what have been anticipated (3.0133), but one should note that 3.15 corresponds only to superconducting behavior without considering the normal state behavior. Therefore, by considering only the normal state behavior, decrement of $\rho_c(T)$ with increasing Pr doping only occurs if $z + \delta < 3.0133$. In short, if $3.0133 < z + \delta < 3.15$, then the materials are superconductors for all x doping but $d\rho_c(T)/dT$ changes from positive to negative with x. However, if $z + \delta < 3.0133$ then the materials are also superconductors for all doping, x but $d\rho_c(T)/dT$ remains positive and $\rho_c(T)$ will decrease with x. Finally, if $z + \delta > 3.15$, then

3

the materials are superconductors for only upto a certain x doping in which, $d\rho_c(T)/dT$ changes from positive to negative and $\rho_c(T)$ would increase with x. Note that the prediction above is only valid for very pure materials without any significant impurity phases. $\rho_c(T)$ data at high temperatures [31] surprisingly decreases with further Pr doping from $x = 0.42$ to 0.55 though it remains semiconducting. This phenomenon could be possibly due to variation in the magnitude of $z + \delta$ where the value for $z + \delta$ might change from; $z + \delta$ (at $x = 0.42$) $> z + \delta$ (at $x = 0.53$) $> z + \delta$ (at $x = 0.55$). I.e., $Pr^{z+\delta}$ varies with doping or concentration, x. Apart from that, as evidenced in experimental data, the phenomenological ab-plane resistivity model (Eq. 9) indicates a linear relationship between temperature and in-plane resistivity due to gapless nature of charge carriers, thus, pointing towards an anomalous Fermi liquid behavior within Cu-O_2 planes.

IV. CONCLUSIONS

Two-dimensional Fermion characteristics have been employed to derive charge carriers' concentration. Anomalous gap parameter (E_I) and charge carriers concentration that has been estimated via best fitting for all superconducting samples of polycrystalline and crystalline are in the order of 4-9 meV and 10^{16} m^{-2} respectively. As a matter of fact, variations that occurred in $\sqrt{np}$, A and E_I are due to variations in sample's purity, methods of preparation and its compositions. Phenomenological ab-plane resistivity model (Eq. 9) indicates a linear relationship between temperature and in-plane resistivity due to gapless nature of charge carriers, signifying an anomalous Fermi liquid behavior in accordance with nested Fermi liquid theory. In addition, disorder of valence state in certain elements such as Pr^{3+}, Pr^{4+} in term of average E_I also contributes to the normal state behavior such as from metallic-like to semiconducting-like behaviors. In other words, $d\rho_c(T)/dT$'s varies from positive to negative or vice versa with selected doping or substitutions at certain temperature. Simply put, polycrystalline resistivity's slope is dependent on average ionization energy, where valence state of Pr which is above 3+ could be estimated from $d\rho_c(T)/dT$ transition upon substitution of Pr into Y sites as Eq. 11 states.

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TABLE I. Calculated values of the temperature independent parameter (A), anomalous gap parameter (E_I), resistivity slope ($d\rho(T)/dT$) and the concentration of charge carriers ($\sqrt{np}$) at 300 K and 150 K in the normal state of high-T_c superconducting compounds. Note that the magnitudes of E_I are in the range of 4-9 meV and the charge carriers' concentration is determined to be in the order of 10^{16} via appropriate fittings. $d\rho(T)/dT$ is calculated from the experimental $\rho_{poly}(T)$ plots. Note that $\sqrt{np}$ is obtained from Eq. 7. k_B is the Boltzmann constant.

FIG. 1. The resistivity curves of crystalline $YBa_2Cu_3O_7$ (Sample B: with volume of unit cell = $630 \times 640 \times 75$ μm) of 123 phase were calculated using Eq. 10. Both $\rho_c(T)$ and $\rho_{ab}(T)$ were computed using Montgomery's algorithm. The single crystals (A and B) were grown by a flux method based on BaO. The ratio a/c for sample B is approximately 8.

FIG. 2. The resistivity of the crystalline $YBa_2Cu_3O_7$ (Sample C: with volume of unit cell = $390 \times 400 \times 25$ μm) of 123 phase were obtained by utilizing Eq. 10. Both $\rho_c(T)$ and $\rho_{ab}(T)$ were computed using Montgomery's algorithm. The ratio a/c for sample C is approximately 16.

FIGURE 1
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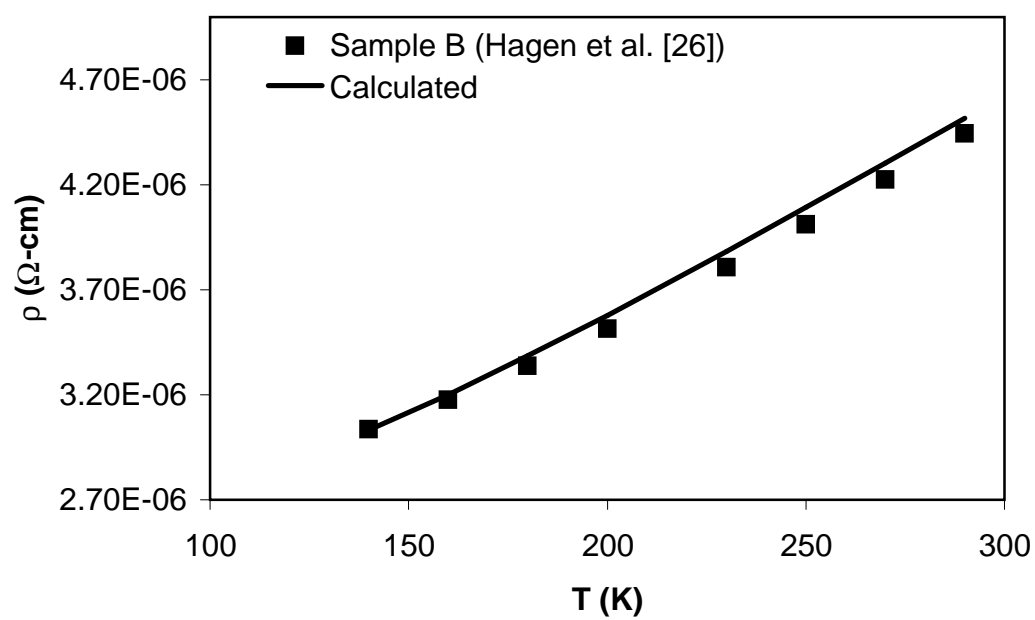
Table 1
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Samples, [Ref.]	A	E_I [J]	E_I [meV]	$\sqrt{np}$ [m^{-2}] 300 K $\times 10^{16}$	$\sqrt{np}$ [m^{-2}] 150 K $\times 10^{16}$	d$\rho(T)$ / dT [$\mu\Omega$-cmK^{-1}]
YBa$_2$Cu$_3$O$_7$ Sample B, [26]	1.1458×10^{-8}	$89.2k_B$	7.69	8.022	5.959	-
YBa$_2$Cu$_3$O$_7$ Sample C, [26]	1.1268×10^{-8}	$94.0k_B$	8.11	7.895	5.771	-
GaSr$_2$(Y$_{0.6}$Ca$_{0.4}$)Cu$_2$O$_7$, [27]	1.3002×10^{-5}	$69.3k_B$	5.98	8.572	6.804	~0
YSrBaCu$_3$O$_7$ Sample [O], [13]	1.1712×10^{-6}	$53.5k_B$	4.61	9.036	7.560	1.10
Y$_{0.8}$Pr$_{0.2}$SrBaCu$_3$O$_7$ Sample [O], [13]	1.8589×10^{-6}	$76.6k_B$	6.61	8.366	6.481	1.60
YBa$_2$Cu$_3$O$_x$, [28]	5.5747×10^{-6}	$95.6k_B$	8.25	7.853	5.710	4.25
YBa$_2$Cu$_3$O$_{7-x}$ Sample A, [29]	6.3767×10^{-6}	$92.0k_B$	7.94	7.948	5.849	5.00
Y(Ba$_{0.85}$Pb$_{0.15}$)$_2$Cu$_3$O$_{7-x}$ Sample B, [29]	5.4926×10^{-6}	$79.8k_B$	6.88	8.278	6.344	4.90
Tl(Sr$_{1.6}$Sm$_{0.4}$)CaCu$_2$O$_{7-x}$, [30]	2.8695×10^{-5}	$59.5k_B$	5.13	8.857	7.264	2.60
Tl(Sr$_{1.6}$Dy$_{0.4}$)CaCu$_2$O$_{7-x}$, [30]	2.2192×10^{-5}	$66.2k_B$	5.71	8.661	6.946	2.00

FIGURE 2
A.D.Arulsamy